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Evaluation of the Temperature-Dependent Adsorption and Corrosion Inhibition Performance of *Rhizophora racemosa* and *Rhizophora harrisonii* Leaf Extract on Mild Steel in Nitric Acid Medium

*Odoronam J. F., Ewa, B. A., Abbas-Adediran, H., Arowosegbe, W. P.

National Biotechnology Research and Development Agency, Abuja

Corresponding Author: odoronamjoy@gmail.com; 08034275403

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ABSTRACT

The search for environmentally sustainable corrosion inhibitors has intensified due to the environmental and health concerns associated with conventional synthetic inhibitors. This study evaluated the influence of temperature on the adsorption characteristics and corrosion-inhibition performance of Rhizophora racemosa and Rhizophora harrisonii leaf extracts on mild steel in 1 M nitric acid solution using the weight-loss technique. The effects of inhibitor concentration (0.4, 0.6, 0.8, and 1.0 g/L), temperature (303.15-343.15 K), and immersion time (2-10 hours) were investigated. The corrosion inhibition efficiency of both extracts improved with increasing inhibitor concentration, demonstrating enhanced adsorption of phytochemical constituents onto the mild steel surface. Temperature-dependent adsorption behaviour was evaluated using Langmuir and Freundlich adsorption isotherms. The adsorption pattern of Rhizophora harrisonii conformed predominantly to the Langmuir model at elevated temperatures. In contrast, Freundlich behaviour was more evident at lower temperatures, indicating changes in the adsorption mechanism with temperature. Activation energies of 17.6 kJ mol^{-1} for Rhizophora racemosa and 21.8 kJ mol^{-1} for Rhizophora harrisonii confirmed a physisorption mechanism. These findings show that both mangrove leaf extracts provide effective, eco-friendly corrosion protection for mild steel in nitric acid and have considerable potential as sustainable green corrosion inhibitors.

Key words: Corrosion inhibition, adsorption isotherms, thermodynamic parameters.

INTRODUCTION

Mild steel is one of the most extensively utilised engineering materials because of its excellent mechanical strength, ease of fabrication, low cost, and wide range of industrial applications. It is commonly employed in the construction, petrochemical, food processing, energy, transportation, and manufacturing industries (Parthipan *et al.*, 2018). Despite these advantages, mild steel is highly susceptible to corrosion in aggressive environments (Goyal *et al.*, 2018), particularly acidic media used in industrial processes such as acid pickling, descaling, cleaning, and chemical production.

Corrosion is a naturally occurring electrochemical process in which metals gradually deteriorate through interaction with the surrounding environment (Goyal *et al.*, 2018). The degradation of metallic materials not only reduces their service life but also results in significant economic losses, operational downtime, safety risks, and increased maintenance costs (Popoola *et al.*, 2013). In acidic environments, corrosion proceeds through anodic dissolution of iron accompanied by cathodic reduction reactions, leading to the formation of corrosion products that progressively weaken the metal's structural integrity (Brown *et al.*, 2018). Consequently, effective corrosion control remains an important challenge across many industrial sectors.

The electrochemical corrosion of iron involves oxidation at anodic sites and oxygen reduction at cathodic sites (Brown *et al.*, 2018). These reactions ultimately produce hydrated iron oxides responsible for rust formation. The following reactions can represent the process:

$Fe(s) + 2H_2O \rightarrow Fe^{2+}(aq) + 2e^-$----------(1)
$O_2(g) + 2H_2O(l) + 4e^-$ à 4OH_(aq)--------(2)
$Fe^{2+} + 2OH^-$ à $Fe(OH)_2$)-------(3)
$4Fe(OH)_2 + O2 + 2H_2O \rightarrow 4Fe(OH)_3$(4)

Among the various methods developed to minimise corrosion, applying corrosion inhibitors has proven to be one of the most practical, economical, and efficient approaches (Ladan *et al*., 2017). Corrosion inhibitors reduce the rate of metal dissolution by adsorbing onto the metal surface and forming a protective barrier that limits contact between the corrosive medium and the substrate (Kadhim *et al*., 2021). Although synthetic inhibitors generally provide excellent protection, many contain toxic substances such as chromates and phosphates that present serious environmental and health concerns, including carcinogenic effects following prolonged exposure (Loto *et al*., 2015). Increasing environmental regulations and the growing demand for sustainable industrial practices have therefore stimulated research into naturally occurring, biodegradable, and environmentally friendly corrosion inhibitors.

Plant-derived extracts have attracted considerable attention as green corrosion inhibitors because they are renewable, inexpensive, biodegradable, and rich in naturally occurring phytochemicals that can interact with metal surfaces. Bioactive compounds, including tannins, flavonoids, alkaloids, saponins, and polyphenols, contain heteroatoms and aromatic structures that facilitate adsorption onto metallic substrates, thereby reducing corrosion rates (Patricia *et al*., 2017). These naturally occurring compounds offer an environmentally acceptable alternative to conventional inorganic inhibitors while maintaining effective corrosion protection.

Rhizophora racemosa and *Rhizophora harrisonii* are two important red mangrove species belonging to the Rhizophoraceae family. These species are widely distributed along the West African coastline and are abundant within the mangrove ecosystems of the Niger Delta, Nigeria. In addition to their ecological importance, mangrove plants are rich sources of tannins and other phenolic constituents that possess strong adsorption characteristics, making them promising candidates for green corrosion inhibition (Brycki *et al.,* 2018). For the present investigation, fresh leaves of both species were collected from the Pennington River in Bayelsa State, Nigeria, where they occur naturally.

Temperature is one of the most influential factors affecting corrosion processes and inhibitor performance because it alters both the rate of metal dissolution and the adsorption behaviour of inhibitor molecules (Chadili *et al*., 2021). Understanding the relationship between temperature and adsorption is therefore essential for evaluating the effectiveness of plant-based inhibitors under varying operating conditions. Adsorption isotherm models, together with thermodynamic parameters such as activation energy, Gibbs free energy, enthalpy, and entropy, provide valuable information regarding the mechanism, spontaneity, and nature of inhibitor adsorption on metal surfaces.

The present study therefore evaluates the temperature-dependent adsorption characteristics and corrosion inhibition performance of *Rhizophora racemosa* and *Rhizophora harrisonii* leaf extracts on mild steel in 1 M nitric acid solution using the weight loss technique. The influence of inhibitor concentration and temperature on corrosion behaviour was investigated, and Langmuir and Freundlich adsorption isotherms, along with thermodynamic analyses, were used to elucidate the adsorption mechanism responsible for the observed corrosion protection.

MATERIALS AND METHODS

Plant Materials

Fresh leaves of *Rhizophora racemosa* and *Rhizophora harrisonii* were collected from the Pennington River in Southern Ijaw Local Government Area of Bayelsa State, Nigeria. The plant materials were authenticated based on their morphological characteristics before being processed for extraction.

Preparation of Leaf Extracts and Inhibitor Solutions.

The collected leaves were thoroughly washed with running tap water to remove adhering dirt and then rinsed with distilled water. The cleaned samples were sun-dried for 72 hours, then oven-dried at 105°C for 48 hours to remove residual moisture. The dried leaves were ground into a fine powder.
The powdered samples were immersed separately in absolute ethanol and allowed to stand for 72 hours to facilitate solvent extraction of the phytochemical constituents. The resulting mixtures were decanted, and the filtrates were concentrated in a water bath maintained at 70°C until semi-solid extracts were obtained. The extracts were transferred into clean sample bottles, sealed properly, and stored until required for the corrosion studies.

Working inhibitor solutions having concentrations of 0.4, 0.6, 0.8, and 1.0 g/L were prepared by dissolving the appropriate quantities of each extract in 1 M nitric acid (HNO_3) solution.

Preparation of Mild Steel Specimens

Mild steel sheets were mechanically cut into 5 cm × 4 cm coupons. The coupon surfaces were polished successively with different grades of silicon carbide abrasive paper to obtain smooth, uniform surfaces. After polishing, the specimens were rinsed with distilled water, degreased with ethanol, air-dried, and stored in a desiccator before use.

Preparation of the Corrosive Medium

The corrosive medium consisted of 1 M nitric acid (HNO_3) prepared by appropriate dilution of analytical-grade nitric acid with distilled water.

Weight Loss Measurements

The corrosion inhibition performance of the leaf extracts was evaluated using the gravimetric (weight loss) method. Mild steel coupons were immersed in 100 mL of 1 M nitric acid solution containing inhibitor concentrations of 0 (blank), 0.4, 0.6, 0.8, and 1.0 g/L.
The experiments were conducted at temperatures of 303.15, 313.15, 323.15, 333.15, and 343.15 K. The initial weight of each coupon was determined using an analytical balance before immersion. The test solutions were placed in appropriately labelled beakers and maintained at the designated temperatures throughout the experiment.
Coupons were withdrawn after 2, 4, 6, 8, and 10 hours of exposure. After each immersion period, the specimens were rinsed thoroughly with water, cleaned with acetone, air-dried, and reweighed to determine the weight loss. The uninhibited acid solution served as the control experiment under identical conditions.

Determination of Corrosion Parameters

The corrosion rate, inhibition efficiency, and surface coverage were calculated from the measured weight loss values using Equations (5)–(10).

The corrosion rate (CR), expressed in millimetres per year, was calculated using Equation (5):
Corrosion rate CR = ……..(5)
Where:
W1 = Initial weight
W2 = Final weight
T = Immersion time in hours
A = Specimen area in cm^2
D =Specimen density in g/cm3;
CR = Corrosion rate in millimetres per year.

Inhibitor Efficiency Calculation: ……(6)
Where Wo is the weight reduction without inhibitor, and Wi is the weight loss with inhibitor.

Surface Coverage (θ) Calculation: ……..(7)
Where:
Wo = weight loss without inhibitor
Wi = weight loss with inhibitor
Adsorption Isotherm Studies:

The adsorption behaviour was analysed using the Langmuir and Freundlich isotherm models using equation 8-.
Langmuir Isotherm ----------(8)
Where : C= inhibitor concentration(gL^{-1}) and K_{ads}=adsorption equilibrium constant

Freundlich Isotherm
$\log\Theta=\log K+n\log C$, ----------(9)
Where K and n are Freundlich constants.

The activation energy was calculated using the Arrhenius equation:

$\ln CR = \ln A - \frac{Ea}{RT}$ ----------(10)

Where CR=Corrosion rate (g $cm^{-2}h^{-1}$, A = Arrhenius constant, EA = Activation Energy (KJ mol^{-1}), R is the Universal gas constant = (8.314J $mol^{-1}K^{-1}$), T=Absolute Temperature (Kelvin)

The Gibbs Free Energy of Adsorption was calculated using the formula:

$DG^{o}_{ads} = -RT \ln(55.5K_{ads})$

Where DG^{o}_{ads} = Standard Gibbs free energy of adsorption (J/mol or KJ/mol), R= 8.314J $mol^{-1}K^{-1}$ Universal gas constant, K_{ads} = Adsorption equilibrium constant and 55.5 = molar concentration of water in solution (mol/L

RESULTS AND DISCUSSION

Effect of Inhibitor Concentration

The experimental results indicate that increasing the concentration of both leaf extracts enhanced corrosion inhibition performance across the temperature range investigated. This trend shows that higher concentrations provide greater surface coverage of mild steel by the active phytochemical constituents, reducing direct interaction between the metal surface and the corrosive medium (Augustine *et al.,* 2018). For *Rhizophora racemosa*, inhibition efficiency increased progressively with concentration at each experimental temperature. At 303.15 K, the inhibition efficiency rose from 4.50% at 0.4 g/L to 21.51% at 1.0 g/L. Similar concentration-dependent improvements were observed at 313.15 K and 323.15 K. The highest inhibition efficiency (81.65%) was achieved at 333.15 K with an inhibitor concentration of 1.0 g/L, indicating optimal protective performance under these conditions. Inhibition efficiency slightly decreased at 343.15 K, suggesting that higher temperatures may reduce the stability of the adsorbed protective film (Chadili *et al*., 2021; Menaka & Subhashin, 2016; Deyab *et al*., 2015).

Table 1: Temperature and Concentration Effect on Inhibition Efficiency of *Rhizophora racemosa* for Nitrate acid (HNO_3).

Temperature (K)	Concentration (g/L)	Reduction in Weight (g)	Inhibition Efficiency (%)	Corrosion Rate (g/cm^2/hr)	Surface Coverage (θ)
303.15	Blank	2.09		0.1173	
	0.4	2.00	4.50	0.1122	0.043
	0.6	1.95	7.18	0.1094	0.067
	0.8	1.84	13.58	0.1032	0.120
	1.0	1.72	21.51	0.0965	0.177
313.15	Blank	2.23		0.1251	
	0.4	2.07	7.73	0.1161	0.072
	0.6	2.01	10.95	0.1128	0.099
	0.8	1.95	14.36	0.1094	0.126
	1.0	1.88	18.62	0.1055	0.157
323.15	Blank	2.48		0.1391	
	0.4	2.20	12.73	0.1234	0.113
	0.6	2.17	14.29	0.1217	0.125
	0.8	2.07	19.81	0.1161	0.165
	1.0	1.98	25.25	0.1111	0.208
333.15	Blank	5.74		0.3220	
	0.4	5.15	11.46	0.2889	0.103
	0.6	4.28	34.11	0.2401	0.254
	0.8	3.55	61.69	0.1992	0.381
	1.0	3.16	81.65	0.1773	0.449
343.15	Blank	6.06		0.3399	
	0.4	5.55	9.19	0.3114	0.084
	0.6	5.11	18.59	0.2867	0.157
	0.8	4.97	21.93	0.2788	0.180
	1.0	4.87	24.44	0.2732	0.196

A comparable trend was obtained for *Rhizophora harrisonii*. At 303.15 K, inhibition efficiency increased from 2.96% at 0.4 g/L to 35.71% at 1.0 g/L. As the concentration increased, the corrosion rate decreased while surface coverage increased, confirming more effective adsorption of inhibitor molecules onto the mild steel surface (Augustine *et al.,* 2018). Maximum inhibition efficiency (84.57%) was likewise obtained at 333.15 K and 1.0 g/L, demonstrating that *Rhizophora harrisonii* exhibited slightly superior inhibition performance compared with *Rhizophora racemosa* under identical experimental conditions. The observed concentration-dependent inhibition behaviour may be attributed to the increasing availability of naturally occurring phytochemicals such as tannins, flavonoids, and polyphenol compounds in the extract. These constituents adsorb onto the mild steel surface and form a protective barrier that limits acid attack and suppresses metal dissolution, consistent with findings reported by Augustine et al. (2018).

Table 2: Temperature and Concentration Effect on Inhibition Efficiency of *Rhizophora harrisonii* for Nitrate acid (HNO_3).

Temperature (K)	Concentration (g/L)	Reduction in Weight (g)	Inhibition Efficiency (%)	Corrosion Rate ($g/cm^2/hr$)	Surface Coverage (θ)
303.15	Blank	2.09		0.1173	
	0.4	2.03	2.96	0.1139	0.029
	0.6	1.83	14.21	0.1027	0.124
	0.8	1.76	18.75	0.0987	0.158
	1.0	1.54	35.71	0.0864	0.263
313.15	Blank	2.23		0.1251	
	0.4	2.18	2.29	0.1223	0.022
	0.6	2.11	5.69	0.1184	0.054
	0.8	2.05	8.78	0.1150	0.081
	1.0	2.00	11.50	0.1122	0.103
323.15	Blank	2.48		0.1391	
	0.4	2.32	6.90	0.1301	0.067
	0.6	2.29	8.30	0.1285	0.O77
	0.8	2.23	11.21	0.1251	0.101
	1.0	2.20	12.73	0.1234	0.113
333.15	Blank	5.74		0.3220	
	0.4	4.06	41.38	0.2278	0.293
	0.6	3.77	52.25	0.2115	0.343
	0.8	3.26	76.07	0.1829	0.432
	1.0	3.11	84.57	0.1745	0.458
343.15	Blank	6.06		0.3400	
	0.4	5.99	1.17	0.3360	0.012
	0.6	5.91	2.54	0.3316	0.025
	0.8	5.88	3.06	0.3299	0.030
	1.0	5.02	20.72	0.2816	0.172

Table 1 and Table 2 above present the influence of inhibitor concentration and temperature on the corrosion behaviour of mild steel in 1 M nitric acid containing *Rhizophora racemosa* and *Rhizophora harrisonii* leaf extracts, respectively.

Effect of Temperature

Temperature exerted a considerable influence on both the corrosion rate and the inhibition efficiency of the investigated extracts. In the uninhibited (blank) solution, corrosion rate increased steadily as temperature increased, indicating accelerated dissolution of mild steel in nitric acid at elevated temperatures. In the inhibited systems, corrosion rates also increased with increasing temperature; however, the presence of both leaf extracts significantly reduced metal dissolution compared with the blank solution. The greatest inhibition efficiencies for both extracts occurred at 333.15 K, suggesting that moderate temperature elevation enhanced adsorption of the inhibitor molecules and promoted formation of a more compact protective layer on the mild steel surface.

Beyond this optimum temperature, inhibition efficiency declined slightly at 343.15 K. This pattern likely

reflects partial desorption of adsorbed inhibitor molecules due to increased thermal agitation, exposing more active sites on the metal surface to the corrosive medium. Similar temperature-dependent adsorption behaviour has been reported by Chadili et al. (2021), Menaka and Subhashini (2016), and Deyab et al. (2015). Overall, the results indicate that although corrosion becomes more aggressive at higher temperatures, both mangrove leaf extracts maintain appreciable inhibitory activity across the investigated temperature range.

Adsorption Isotherm Studies

Adsorption isotherm analysis was employed to describe the interaction between inhibitor molecules and the mild steel surface. The adsorption pattern varied with temperature, indicating that temperature influenced both the extent and mode of adsorption. For *Rhizophora harrisonii*, the adsorption data exhibited better agreement with the Freundlich adsorption isotherm at lower temperatures (303.15-313.15 K), suggesting heterogeneous surface adsorption involving multiple adsorption sites. As temperature increased to 323.15-333.15 K, the adsorption became more consistent with the Langmuir isotherm, indicating the formation of a predominantly monolayer protective film on relatively uniform adsorption sites (Adamu et al., 2024). At 343.15 K, deviation from the ideal Langmuir pattern was observed, probably due to partial desorption of adsorbed molecules resulting from increased thermal energy (Chadili *et al.*, 2021).

The adsorption characteristics of *Rhizophora racemosa* also demonstrated improved conformity with the Langmuir adsorption model at elevated temperatures, indicating relatively uniform adsorption of inhibitor molecules on the mild steel surface. These observations are consistent with previous reports that adsorption behaviour of plant-derived corrosion inhibitors depends strongly on temperature and surface interactions (Adamu et al., 2025).

Thermodynamic Parameters

Thermodynamic analysis provided further insight into the corrosion inhibition mechanism of the investigated leaf extracts. The activation energy values calculated from the Arrhenius relationship were 17.6 kJ mol^{-1} for *Rhizophora racemosa* and 21.8 kJ mol^{-1} for *Rhizophora harrisonii.* These relatively low activation energies indicate that inhibition occurs predominantly through physical adsorption (Chaoui *et al.*, 2023).

The Adsorption Equilibrium Constant (Kads) increased with temperature, indicating enhanced adsorption of the mangrove extract molecules on the mild steel surface. The calculated Gibbs free energy values (−6.56 and −11.27 kJ/mol) suggest that the adsorption process is spontaneous and predominantly physical. The positive enthalpy values confirm the endothermic nature of adsorption, while positive entropy values suggest increased disorder at the metal-solution interface during inhibitor adsorption. In general, both mangrove extracts act through spontaneous, endothermic, and predominantly physical adsorption mechanisms.

The spontaneous nature of adsorption (negative ΔG°) suggests favourable interaction between inhibitor molecules and the mild steel surface. The magnitude of ΔG° (< −20 kJ/mol) indicates that the adsorption process is predominantly physical, involving electrostatic interaction rather than chemical bond formation. The higher activation energy in inhibited systems compared to the blank solution further supports the physical adsorption mechanism. The superior performance of *Rhizophora harrisonii* at 333 K may be attributed to a higher concentration of active phytoconstituents, resulting in greater surface coverage.

CONCLUSION

Results from the studies demonstrated that *Rhizophora racemosa* and *Rhizophora harrisonii* leaf extracts are effective, environmentally friendly corrosion inhibitors for mild steel in nitric acid medium. Their high inhibition efficiencies, favourable adsorption characteristics, and environmentally benign nature highlight their potential as sustainable alternatives to conventional synthetic corrosion inhibitors in industrial applications.

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